Impact of mercury emissions from historic gold and silver mining: Global modeling

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ARTICLE INFO

ABSTRACT

We compare a global model of mercury to sediment core records to constrain mercury emissions from the 19th century North American gold and silver mining. We use information on gold and silver production, the ratio of mercury lost to precious metal produced, and the fraction of mercury lost to the atmosphere to calculate an a priory mining inventory for the 1870s, when the historical gold rush was at its highest. The resulting global mining emissions are 1630 Mg yr^{-1}, consistent with previously published studies. Using this a priori estimate, we find that our 1880 simulation over-predicts the mercury deposition enhancements archived in lake sediment records. Reducing the mining emissions to 820 Mg yr^{-1} improves agreement with observations, and leads to a 30% enhancement in global deposition in 1880 compared to the pre-industrial period. For North America, where 83% of the mining emissions are located, deposition increases by 60%. While our lower emissions of atmospheric mercury leads to a smaller impact of the North American gold rush on global mercury deposition than previously estimated, it also implies that a larger fraction of the mercury used in extracting precious metals could have been directly lost to local soils and watersheds.

1. Introduction

Gold and silver mining in North, Central, and South America, and in Australia, which used mercury amalgamation to extract the precious metals, is estimated to have released approximately 156,000 Mg of mercury into the atmosphere and over 250,000 Mg of mercury into the environment between 1580 and 1900 (Nriagu, 1994). The North American gold and silver rushes that began when gold was discovered in California in 1847 represent a particularly intense period of mining. Silver production rose from 1.2 Mg yr^{-1} in 1850 to 940 Mg yr^{-1} in 1880 (Bureau of the Census, 1989). From 1850 to 1900 atmospheric mercury emissions from gold and silver mining in the United States averaged 780 Mg yr^{-1} (Nriagu, 1994), compared to present day U.S. anthropogenic emissions of approximately 100 Mg yr^{-1} (Pacyna et al., 2006). Because mercury is both toxic and persistent in the environment, this mercury release is still a concern today. Sites contaminated with mercury during the historic gold rush continue to cause mercury contamination in California (Alpers et al., 2005) and Nevada (Wayne et al., 1996) watersheds.

Sediment cores provide a record of changes in mercury deposition through history. Lake sediments suggest that modern mercury deposition in the northern hemisphere is 2–4 times larger than the pre-industrial background value (Lindberg et al., 2007; Biester et al., 2007). For the historic North American gold rush period, however, the record is less clear. Schuster et al. (2002) found a factor of 5 enhancement in total mercury concentration and deposition in a Wyoming glacier ice core layer corresponding to the late 19th century, which they attribute to gold-mining emissions. In contrast, Lamborg et al. (2002) did not find a clear mining signal in lake sediments from Nova Scotia and New Zealand, and suggest that mercury from historic mining remained close to its source rather than being deposited globally.

Modeling studies provide insight into the relationship between mining emissions of mercury and the deposition changes recorded in sediment core record. Hudson et al. (1995) included mercury emissions of 2200 Mg yr^{-1} at the peak of the North American gold rush in a pre-technological to modern box-model simulation of the mercury cycle. Compared to lake sediments from the upper Midwest United States (Swain et al., 1992), these mining emissions result in too large a peak in mercury deposition around 1880 (Hudson et al., 1995). Pirrone et al. (1998) estimate that North American mercury emissions peaked at 1708 Mg yr^{-1} in 1879

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mostly due to mining emissions, but do not find a corresponding enhancement in this period in sediment cores from the Great Lakes.

In this study, we estimate mercury emissions from the North American gold rush era based on records of gold and silver production. We then use the GEOS-Chem global atmosphere–ocean–land mercury model to calculate the global impact of these emissions on deposition. Finally, we examine the consistency between our estimates and historic core records from around the world.

2. Methods

2.1. Historic core records

Numerous studies have used lake sediment, peat bog, or ice cores to interpret the history of mercury deposition and infer the anthropogenic enhancement ratio (ER) given by the ratio of modern to pre-industrial accumulation rates ($ER_{modern/p-i}$). Several uncertainties are important in relating these records to atmospheric deposition, including variability in sedimentation rates (Engstrom and Wright, 1984) and sediment focusing (Perry et al., 2005), post-deposition mobility of mercury within the core (Gobeil and Cossa, 1993), and the contribution of mercury from the catchment area rather than from atmospheric deposition (Swain et al., 1992). For this study, we select cores from the literature that report mercury as accumulation rate rather than concentration to reduce the influence of variable sediment flux, and we exclude studies that report significant post-depositional redistribution of mercury. Since we are comparing with a global model, we use cores from areas where atmospheric deposition is expected to dominate over runoff from local sources.

The long lifetime of total gaseous mercury (>6 months) implies that large emissions from the North American gold rush mining era should have affected deposition globally, and not just in North America. We thus assemble a global dataset including sediment core records from North America (Engstrom and Swain, 1997; Fitzgerald et al., 2005; Kamman and Engstrom, 2002; Lamborg et al., 2002; Landers et al., 1998, 2008; Lorey and Driscoll, 1999; Swain et al., 1992), South America (Lacerda et al., 1999), Greenland (Asmund and Nielsen, 2000; Bindler et al., 2001), Europe (Verta et al., 1989), Siberia (Landers et al., 1998), and New Zealand (Lamborg et al., 2002). For greater geographic coverage, we also include observations from an ice core in North America (Schuster et al., 2002), and two peat bog records in South America (Biester et al., 2002) and Europe (Roos-Barraclough and Shotyk, 2003).

As some of the cores do not extend back to pre-industrial times and some lack the temporal resolution to provide an 1880 value, we define 3 enhancement ratios: $ER_{modern/p-i}$, $ER_{1880/p-i}$, and $ER_{1880/modern}$. $ER_{1880/p-i}$ is the product of the other two ratios. Modern is defined as the sediment data point closest to the year 2000; p-i represents the pre-industrial period, which includes data from before 1840; and 1880 is the date chosen to represent the North American gold and silver rush. If multiple cores lie in the same model grid box or within 2 degrees latitude and longitude of each other, we average them together. Table S1 in the supplemental materials shows the dataset used for this study.

2.2. A priori mining emissions

We derive our global a priori mining emission inventory for the 1870s using the following equation:

$$F_{mining} = \left(P_{gold} + P_{silver} \right) \times R_{Hg/metal} \times f_{atmos} \qquad (1)$$

where F_{mining} is the total mass of elemental mercury (Hg^0) released to the atmosphere. P_{gold} is the mass of gold produced, P_{silver} is the

mass of silver produced, $R_{Hg/metal}$ is the mass ratio of mercury lost to gold or silver produced and f_{atmos} is the fraction of mercury released to the atmosphere. Mitchell (2003a,b) reports the mass of gold and silver produced by country for each year. Silver dominates the total production, with the United States producing 760 Mg silver compared to 50 Mg gold in 1875 (Mitchell, 2003b). To determine $P_{gold} + P_{silver}$, we sum Mitchell's (2003a,b) gold and silver production numbers and average from 1870 to 1879 to account for the long atmospheric lifetime of Hg^0 and the temporal resolution of the sediment cores. Mining emissions are distributed evenly across each country, except in the United States, where we assume that the emissions occurred in the western part of the country.

The ratio of mercury lost to gold or silver produced is uncertain. In 18th century South American silver mining, $R_{Hg/metal}$ was estimated to be approximately 2:1, although in some regions the ratio was closer to 1.5:1 or 1:1 and the ratio varied greatly between regions and years depending on the ore and the availability of mercury (Brading and Cross, 1972; Fisher, 1977; Nriagu, 1994). Nriagu (1994) estimate that the ratio was approximately 1:1 in 19th century South and Central America. Pfeiffer and Lacerda (1988) estimated a ratio of 1.3:1 for modern gold miners using mercury amalgamation in Brazil. Other modern estimates usually lie between 1:1 and 1.5:1 (Lacerda, 2003). Given that the published estimates for the ratio of mercury lost to gold and silver overlap, we choose a common value of $R_{Hg/metal} = 1.5:1$ for our a priori 1870s emissions estimate.

Estimates also vary for the fraction of mercury released to the atmosphere during the amalgamation process, f_{atmos}. Pfeiffer and Lacerda (1988) estimated that 55% of mercury from modern Amazon gold mining enters the atmosphere, and Lacerda and Salomons (1991) report that 65%–83% is lost to the atmosphere in this region. In contrast, Swain et al. (2007) estimate that only 30% of the mercury used in small-scale gold mining is released directly to the atmosphere. For historic mining, Nriagu (1994) estimates a 60% loss to the atmosphere, and Pirrone et al. (1998) used this 60% value to estimate emissions from North American gold and silver mining. Following these studies, we set f_{atmos} to 0.6 for our a priori emissions.

2.3. Global model

The GEOS-Chem chemical transport model (Bey et al., 2001) simulates mercury in the atmosphere–ocean–land system (Selin et al., 2007, 2008; Strode et al., 2007). The simulation includes tracers for elemental mercury (Hg^0), divalent mercury (Hg^{II}), and particulate mercury (Hg_p), with both oxidation of Hg^0 to Hg^{II} and in-cloud reduction of Hg^{II} occurring in the atmosphere. We use here model version 7.04 (http://www.harvard.as.edu/chemistry/trop/geos) with updates described in Selin and Jacob (2008).

We conduct model simulations for 3 different sets of mercury emissions: pre-industrial, 1880, and modern day. For each emission scenario, we run the model until it reaches steady state. The model has a horizontal resolution of 4° latitude by 5° longitude, and 30 vertical levels. It is driven by assimilated meteorological fields from the NASA Goddard Earth Observing System (GEOS-4) for 2004 for all simulations so that the ER is not affected by interannual variability in precipitation. We compare the ER values from the cores with modeled deposition enhancement ratios for the same time periods. Comparing ER values rather than absolute deposition rates normalizes out some site-specific factors such as average local precipitation (Biester et al., 2007). Note that modern oxidant concentrations (OH and O_3) were not modified for the pre-industrial and 1880 simulations. Thus, we do not address the effects of changing oxidant concentrations on deposition.